

# Fine-structure splitting reduction of ionized impurity bound exciton in quantum dot

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The ground-state energy and fine-structure splitting of ionized shallow donor impurity-exciton complex in quantum dots are investigated. It is found that fine-structure splitting could be largely reduced by the off-center ionized impurities since the anisotropic shape of exciton envelope function is significantly changed. Anomalous Stark shifts of the ground-state energy and efficient tuning of the fine-structure splitting by the external electric field due to the local electric field produced by the ionized impurities are discussed. The scheme may be useful for the design of the quantum dots-based entangled-photon source.

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Semiconductor quantum dots (QDs) have been demonstrated as one of candidates for the entangled two-photon sources, which make them very attractive for applications in the fields of quantum teleportation and quantum computation.<sup>1,2</sup> Much efforts, e.g., thermal annealing and external field tuning,<sup>3,4,5,6,7,8</sup> have been devoted to the reduction of the fine-structure splitting (FSS) of the intermediate exciton states, since a necessary condition in the proposal for a QD-based source of polarization entangled photon pairs is that the intermediate exciton states for the biexciton radiative decay are energetically degenerate.<sup>9</sup> FSS originated from the anisotropic electron-hole exchange interaction is directly determined by the anisotropic shape of the exciton envelope function. Obviously, exciton envelope function may be strongly influenced by the ionized shallow impurities in QDs, and consequently FSS might be changed. In this letter, we study the ground-state FSS of ionized hydrogenic donor impurity-exciton complex in quantum dots and find that it could be largely reduced by the ionized hydrogenic impurities as well as the external electric field.

Recently, with the advancement of QDs growth and measurement technics, it is possible to optically probe fine-structures of single magnetic impurity-doped semiconductor QDs.<sup>10</sup> In this letter, the ground-state energy and FSS of ionized hydrogenic donor impurity-exciton complex in semiconductor QDs under an external in-plane electric field are investigated, since shallow donor impurity is common and well studied in III-V semiconductors, e.g., Si-doped GaAs. The light-hole and spin-orbit-split  $J = 1/2$  valence bands could be reasonably neglected in the calculations, since the heavy-hole component is dominant in the hole ground state of flat InGaAs QDs<sup>11</sup> and we mainly focus on the exciton ground states. Thus the exciton state is composed of 4 combinations of the valence band and the conduction band, i.e.,  $|X\rangle = \sum_{m,s} \sum_{r_e,r_h} \psi_{ms}(r_e, r_h) a_{c_s r_e}^\dagger a_{v_m r_h} |0\rangle$  where the Wannier function representation of the creation and annihilation operators is used,  $\psi_{ms}(r_e, r_h)$  is the exciton envelope function, and  $m$  and  $s$  are the  $z$ -component of the angular momentum of the heavy-hole valence band and the conduction band, respectively. The eigenvalue

equation for  $\psi_{ms}$  is given as

$$\sum_{m's'r'_e r'_h} [H_1 + V_{\text{ex}}(c_s r_e, v_{m'} r'_h; c_{s'} r'_e, v_m r_h)] \psi_{m's'}(r'_e, r'_h) = E \psi_{ms}(r_e, r_h), \quad (1)$$

with

$$H_1 = \delta_{r_e r'_e} \delta_{r_h r'_h} \delta_{s' s} \delta_{m' m} [H_e + H_h + eF \cdot (r_e - r_h) - \frac{e^2}{\epsilon|r_e - r_h|} + V_{\text{int}}(r_h, r_e, q_j)], \quad (2)$$

where  $H_k = p_k^2/2m_k + U_k(r_k)$  ( $k = e, h$ ) is the single particle Hamiltonian,  $U_h$  ( $U_e$ ) is the confinement potential for the hole (electron),  $F$  is an external in-plane electric field, and  $V_{\text{int}}(r_h, r_e, q_j) = \sum_{j=1}^N (e^2/\epsilon|r_h - q_j| - e^2/\epsilon|r_e - q_j|)$  is the Coulomb interaction between charge carriers and ionized impurity centers,  $q_j = (x_j, y_j)$  is the position of the  $j$ th ionized donor impurity,  $N$  is the total number of ionized donor impurities. Similar to the assumption in Ref. 12, an in-plane anisotropic potential is used in modeling single QDs, i.e.,  $U_{e(h)} = \nu_{e(h)} \theta(b/2 - |y_{e(h)}|) \theta(a/2 - |x_{e(h)}|)$ , where  $a$  and  $b$  are the lateral sizes of QDs, and  $\nu_e$  ( $\nu_h$ ) is the conduction (heavy-hole valence) band offset. Whether the geometric shape of single QDs is rectangular or elliptic will not change the qualitative results of this letter.  $V_{\text{ex}}$  is the electron-hole exchange interaction.<sup>12</sup> The material parameters used in the calculations refer to Ref. 13. The computational procedure is that eigenfunctions of spin-independent  $H_1$  is firstly calculated using diagonalization method with more than 4000 basis sets, and then FSS is obtained by calculating the matrix elements of  $V_{\text{ex}}$  in the basis of eigenfunctions of  $H_1$ .

For simplicity, single ionized donor impurity is located along the  $x$ -axis, and the ionized impurity-exciton complex binding energy  $E_b$  is defined as

$$E_b = E(X) - E(D^+, X), \quad (3)$$

where  $E(X)$  is the exciton ground state energy in QDs and  $E(D^+, X)$  is the ground state energy of the ionized donor impurity-exciton complex in the same QD.<sup>14</sup> In

Fig. 1(a), ground-state energies of single ionized donor impurity-exciton complex without the electron-hole exchange interaction in two kinds of anisotropic QDs are shown as functions of impurity position. For the first kind of QDs (QD1) with  $b = 18.0$  nm and  $a = 20.0$  nm and the donor impurity located at the QD center,  $E_b = 5.44$  meV, while for the second kind of QDs (QD2) with  $b = 16.0$  nm and  $a = 20.0$  nm and the donor impurity located at the QD center,  $E_b = 5.56$  meV. As the impurity position  $x_1$  increases from zero, however, the binding energy initially increases and gets a maximal value at about  $x_1 = 5.0$  nm for both QD1 and QD2 as shown in Fig. 1(a). When  $x_1$  exceeds 5.0 nm,  $E_b$  becomes smaller and gradually approaches zero as  $x_1 \rightarrow \infty$ . In Fig. 1(b), corresponding oscillator strength of the exciton transition is also shown.

Including the exchange interaction  $V_{ex}$ , the exciton ground states are split into bright and dark doublets. FSS of bright doublet is mainly determined by the anisotropic shape of InGaAs QDs.<sup>12</sup> FSS of QD2 with larger shape anisotropy and without impurity ( $66 \mu\text{eV}$ ) is larger than that of QD1 with smaller shape anisotropy and without impurity ( $27 \mu\text{eV}$ ), and calculated value of FSS is well consistent with recent experimental results.<sup>15</sup> When there is an ionized donor impurity present at the QD center, FSS is slightly reduced, relative to the  $N = 0$  case, as shown in Fig. 1(c). Interestingly, reduction of FSS is largely enhanced for the off-center donor impurity. At about  $x_1 = 5.0$  nm, FSS gets a minimal value, i.e.,  $17 \mu\text{eV}$  and  $47 \mu\text{eV}$  for QD1 and QD2, respectively. Moreover, FSS could be further reduced when there are more than one ionized donor impurity. For example, the ground-state FSS of two ionized donor impurities-exciton complex in QD1 is only  $6 \mu\text{eV}$ , with the two donor impurities position  $q_1 = (5.0 \text{ nm}, 1.0 \text{ nm})$  and  $q_2 = (5.0 \text{ nm}, -1.0 \text{ nm})$ .

In Fig. 2(a), ground-state energies without the electron-hole exchange interaction for three different systems, i.e., (i) exciton without impurity, (ii) single ionized donor impurity-exciton complex, and (iii) two ionized donor impurities-exciton complex in QDs, are shown as functions of an external electric field along the  $x$ -axis. The single donor impurity position is  $(5.0 \text{ nm}, 0 \text{ nm})$ , and the two donor impurity positions are  $(5.0 \text{ nm}, 1.0 \text{ nm})$  and  $(5.0 \text{ nm}, -1.0 \text{ nm})$ , respectively. The Stark shift of all three cases could be well approximated by a parametric model

$$\Delta E(F) = \alpha F - \frac{1}{2}\beta F^2 + \dots, \quad (4)$$

where  $\alpha$  and  $\beta$  are actually the exciton dipole and polarizability, respectively, along the external field.<sup>16</sup> For the first case without ionized impurities,  $\alpha$  is zero and the Stark shift could be well fitted by the quadratic term. However,  $\alpha$  becomes nonzero for the second and third case because of the opposite Coulomb interactions between electron-ionized donor and hole-ionized donor. At small external electric field, the Stark shift could be well described by the linear and quadratic terms, while

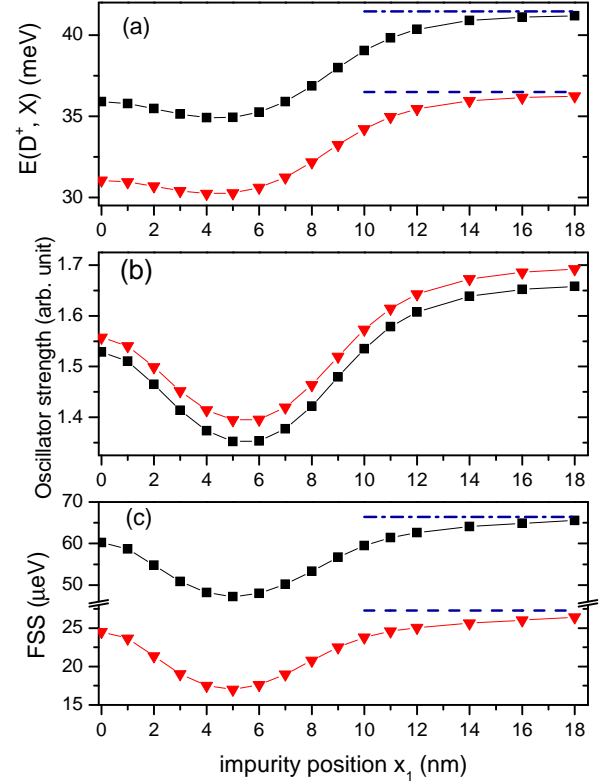


FIG. 1: (Color online) (a) The ground-state energy, (b) corresponding oscillator strength, and (c) fine-structure splitting of single ionized donor impurity-exciton complex as functions of the donor impurity position  $x_1$ , with  $y_1 = 0$  for QD1 (triangles) and QD2 (squares), respectively. The exciton ground-state energy and FSS in QD1 (dash lines) and QD2 (dash-dot lines) without impurities are also shown, respectively.

higher-order terms need to be taken into account at larger external electric field. Oscillator strength of the ground state shows interesting behaviors. For the first case, oscillator strength monotonically decreases as the external electric field, and its behavior is symmetric for the field in the positive and negative directions. For the last two cases, the behavior of the ground-state oscillator strength with the external field is clearly asymmetric. For  $F > 0$ , external field partially counteracts the local field produced by the ionized impurities, and the overlap between electron and hole is initially enhanced. Therefore, the ground-state oscillator strength initially increases. However, as the external field exceeds the effective local field, then oscillator strength is finally reduced as shown in Fig. 2(b). For  $F < 0$ , external field is in the same direction of the effective local field produced by the ionized impurities, and the oscillator strength monotonically decreases.

FSS of the three cases are shown as functions of the external electric field in Fig. 2(c). It can be seen that the behavior of exciton FSS in QDs with off-center ionized

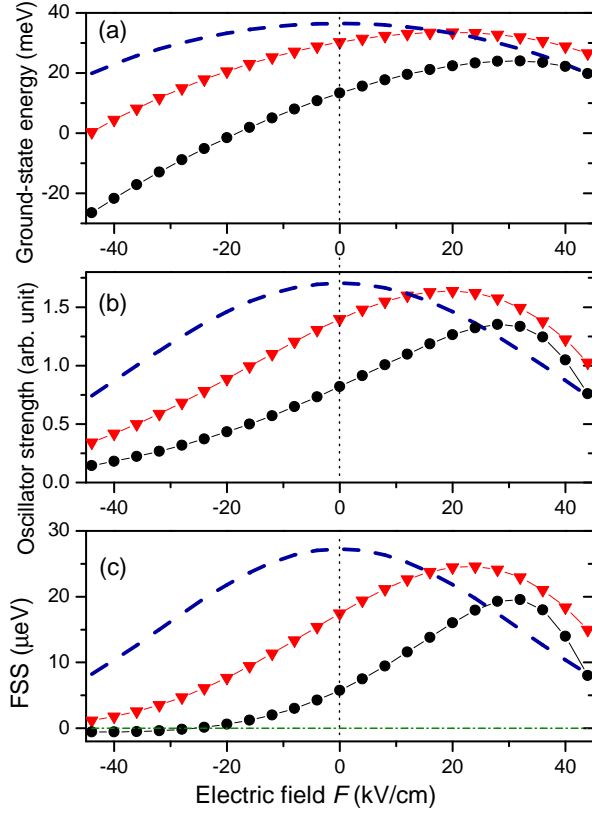


FIG. 2: (Color online) (a) The ground-state energy, (b) corresponding oscillator strength, and (c) FSS in QD1 as functions of the external electric field  $F$  for the following three cases: (i) absence of ionized impurity (dash lines), (ii) one ionized donor impurity (triangles) with  $q_1 = (5.0 \text{ nm}, 0 \text{ nm})$ , (iii) two ionized donor impurities (circles) with  $q_1 = (5.0 \text{ nm}, 1.0 \text{ nm})$ ,  $q_2 = (5.0 \text{ nm}, -1.0 \text{ nm})$ .

impurities is greatly different from that of QDs without ionized impurities. For QDs without ionized impurities, FSS monotonically decreases with the external electric field. However, FSS of QDs with off-center ionized impurities shows asymmetric variations for external electric field in positive and negative directions. We note that FSS shows somewhat similar behaviors as functions of the external electric field with those of the oscillator strength, since both the oscillator strength and FSS are directly related to the overlap between the electron and hole. Interestingly, FSS in the third case could be reduced to less than  $1 \mu\text{eV}$  as  $F < -16 \text{ kV/cm}$ , which is below the typical homogeneous linewidth of the exciton emission lines (determined by the exciton radiative lifetime), and the corresponding oscillator strength at  $F = -16 \text{ kV/cm}$  is about 30% of that in the same QD without ionized impurity and external field. With the help of the local field produced by the off-center donor impurities, FSS in anisotropic QDs could be more efficiently tuned towards zero with external electric field as shown in Fig. 2(c). On the other hand, according to the

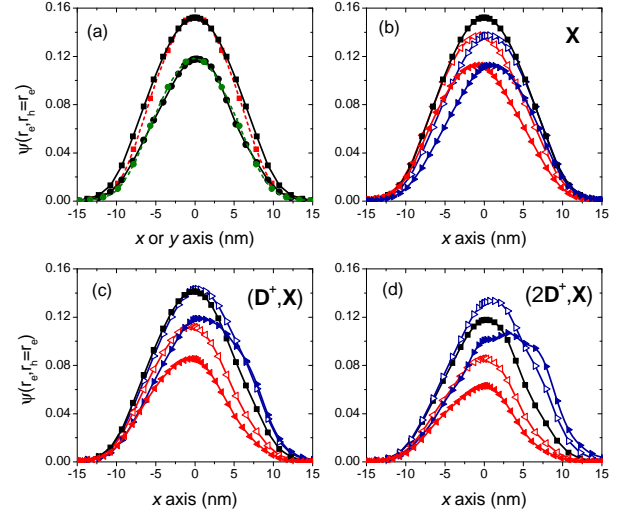


FIG. 3: (Color online) (a)  $\psi(r_e, r_h = r_e)$  along the  $x$ - (solid lines) and  $y$ -axis (dash lines) for the first (squares) and the third case (circles) of Fig. 2, respectively, with  $F = 0 \text{ kV/cm}$ ;  $\psi(r_e, r_h = r_e)$  along the  $x$ -axis of (b) the first, (c) the second, and (d) the third case in Fig. 2, for  $F = -40.0 \text{ kV/cm}$  (leftward triangles),  $-24.0 \text{ kV/cm}$  (open leftward triangles),  $0 \text{ kV/cm}$  (squares),  $+24.0 \text{ kV/cm}$  (open rightward triangles), and  $+40.0 \text{ kV/cm}$  (rightward triangles), respectively.

analysis mentioned above, it is easy to understand that local electric field, produced by ionized impurities, defect trapped electron or hole, and surface charges in real samples could strongly affect the external electric field-tuning of FSS in QDs. Thus the results in this letter might be useful to explain the anomalous external electric-field dependence of FSS observed in the experiment.<sup>8</sup>

In order to better understand the reduction of exciton FSS in QDs with ionized impurities,  $\psi(r_e, r_h = r_e)$  of the ground state in QD1 along the  $x$ - and  $y$ -axis are shown in Fig. 3(a). For QD1 without ionized impurity and external field, the extension of  $\psi(r_e, r_h = r_e)$  along the  $x$ -axis is slightly larger than that along the  $y$ -axis as clearly shown in Fig. 3(a). Thus the electron-hole long range exchange interaction is nonzero and FSS is calculated to be  $27 \mu\text{eV}$ . When there are ionized donor impurities present as in the third case of Fig. 2, both the amplitude and shape of  $\psi(r_e, r_h = r_e)$  are greatly changed due to the local electric field produced by the ionized donor impurities, and the extension along the  $x$ -axis becomes nearly identical to that along the  $y$ -axis as shown in Fig. 3(a). That is why FSS is largely reduced to only  $6 \mu\text{eV}$ . In Figs. 3(b), 3(c), and 3(d),  $\psi(r_e, r_h = r_e)$  of all the three cases in Fig. 2 are shown for several values of the external electric field, and it could be easily seen that both the amplitude and shape of  $\psi(r_e, r_h = r_e)$  are largely changed by the external electric field as well as the ionized donor impurities, which clearly explains the anomalous behaviors of the oscillator strength and FSS in Figs. 2(b) and 2(c).

In summary, we study the ground-state FSS of ionized shallow donor impurities-exciton complex in anisotropic QDs, and find that it could be largely reduced by one or two off-center ionized donor impurities, which strongly influence the exciton envelope functions. Then anomalous Stark shifts and efficient tuning of FSS by the external electric field are clearly shown and discussed. The

study will be helpful and interesting for the research on the QDs-based entangled-photon source.

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